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# TESTING THE PLUTONIUM ISOTOPIC ANALYSIS CODE FRAM WITH VARIOUS CdTe DETECTORS

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## SUMMARY

The isotopic analysis code Fixed-energy Response-function Analysis with Multiple efficiency (FRAM)[1,2] has been proven to successfully analyze plutonium spectra taken with a portable CdTe detector with Peltier cooling, the first results of this kind for a noncryogenic detector.[3] These are the first wide-range plutonium gamma-ray isotopics analysis results obtained with other than Ge spectrometers. The CdTe spectrometer measured small plutonium reference samples in reasonable count times, covering the range from low to high burnup. This paper describes further testing of FRAM with two CdTe detectors of different sizes and resolutions using different analog and digital, portable multichannel analyzers (MCAs).

## I. INTRODUCTION

Nondestructive analysis (NDA) applied to bulk nuclear materials (NMs) is important for nuclear safeguards and material control because of timeliness, cost effectiveness, and containment integrity. Nuclear NDA techniques (calorimetry, neutron coincidence counting) require knowledge of the isotopic composition of the material for quantitative interpretation of the measurements. Gamma-ray spectroscopy with high-resolution detectors is a well developed NDA technique for isotopics.

The use of intrinsic Ge detectors cooled to cryogenic temperatures for isotopic measurements is sometimes difficult or even impossible because of severe access limitations with the sensitive, heavy detectors. Highly portable isotopics measurements are needed for *in-situ* verification of bulk NM quantities or, in many cases, for measurements of holdup quantities.

A new, fully portable CdTe detector with Peltier cooling, developed by Radiant Detector Technologies, LLC[a], and PNPI (Russia), is now offered commercially by Radiant. The detector connects to a power supply that operates under battery power as part of the compact system. The volumes of the CdTe crystals used in the new detectors are approximately 4–10 times smaller than those of CdZnTe crystals with coplanar-grid electrodes. However, the new CdTe detector improves gamma-ray energy resolution by a factor of 4 or 5 compared to CdZnTe. Figure 1 shows the new commercial detector, which adapts recent materials, solid-state advances, cooling technologies, and charge-loss correction techniques.[4,5]

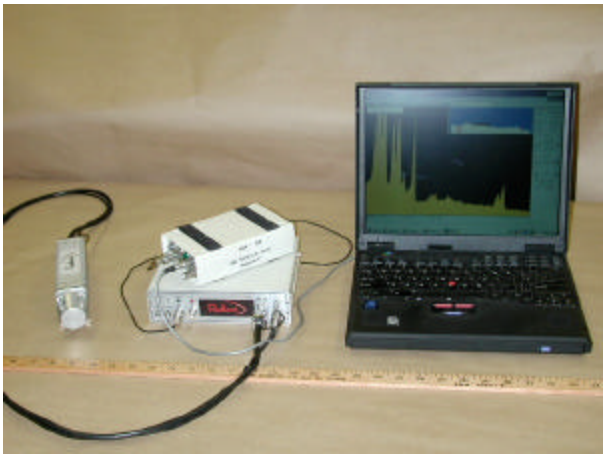

**Figure 1. The Peltier-cooled CdTe detector supplied by Radiant Detector Technologies is at the left. The box at the bottom/center contains Radiant's charge-loss correction unit, a cooling regulator and power supply. The unit at the top/center is the portable MCA-166 from GBS-Elektronik GmbH, Germany.**

This paper describes the gamma-ray measurements with two different CdTe detectors. It presents the detailed results of the wide-range isotopic analysis of plutonium with FRAM v4, the first results of this kind for a noncryogenic detector.

## II. PERFORMANCE: FIRST CdTe DETECTOR

A new, large-area (10 mm x 9 mm x 1.5 mm) Peltier-cooled CdTe detector was the first tested. The cooler is regulated to -40$^{o}$C independent of the ambient temperature. The detector comes with a portable unit that supplies the power to the detector (high voltage and power for the cooler). This unit also corrects for charge loss, the result of the known transport characteristics of CdTe. The analog output pulses from the charge-loss correction circuit are the input to the amplifier or digital signal processor of any MCA system. The operating system is any software that controls the MCA. The charge-loss correction improves spectral resolution without throwing away counts. Figure 2 shows parts of the $^{57}$Co spectrum measured for a fixed count time with digitization of signals: 1) directly from the detector, and 2) from the output of the charge-loss correction circuit. The count rates for these two spectra are the same. Note that the areas under the 122-keV peak are the same, but the peak from the processed spectrum has a much smaller tail and higher peak than the one from the unprocessed spectrum.

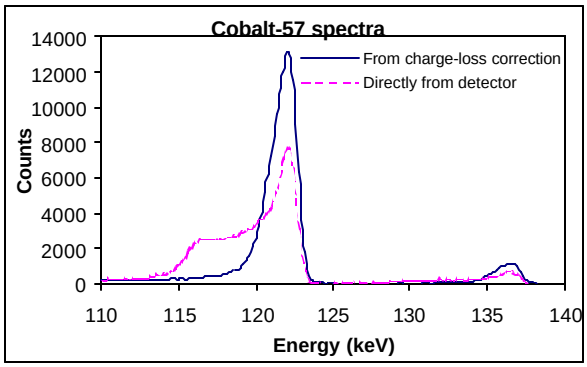


**Figure 2. Spectra of the $^{57}$Co source taken for a fixed count time with the signals directly out of the detector and with the signals from the charge-loss correction amplifier.**

### A. CdTe Compared with Other Detectors

We compared the performance of this CdTe detector with that of three detectors: planar Ge (25-mm diameter and 15-mm depth), NaI (25-mm diameter and 25-mm depth), and coplanar-grid CdZnTe (15 mm x 15 mm x 7 mm). The Ge detector operates at liquid-nitrogen temperature.

Table 1 shows the resolution at 122 keV and 662 keV for these detectors. The count rates for these measurements were about 10 kHz. The resolution of the CdTe detector is roughly triple that of the Ge, 20% of the CdZnTe, and 10% of the NaI detectors.

**Table 1. Resolution (full-width at half maximum) of four detectors using $^{57}$Co and $^{137}$Cs sources**

| Peak energy | NaI | CdZnTe | CdTe | Ge |
|---|---|---|---|---|
| 122-keV | 15.1 | 7.7 | 1.8 | 0.52 |
| 662-keV | 50.6 | 19.9 | 3.8 | 1.27 |

A $^{133}$Ba source was a reference for comparing the efficiencies of the four detectors. The source position was 28 cm from the detector's face in each case. The measured efficiency is based on total counts and the peak areas of the 81-keV and 356-keV peaks from the spectra taken with the four detectors. Table 2 shows the relative efficiencies normalized to that of the CdTe detector.

**Table 2. Relative efficiency of the detectors, normalized to that of CdTe**

| Detector | Total counts | 81-keV | 356-keV |
|---|---|---|---|
| NaI | 33.7 | 16.0 | 85.3 |
| CdZnTe | 7.8 | 3.4 | 20.1 |
| CdTe | 1.0 | 1.0 | 1.0 |
| Ge | 25.8 | 14.2 | 46.6 |

The efficiency at the lower energy is roughly proportional to a detector's active area. The detector thickness is crucial at the higher energy. The CdTe crystal has the smallest area and thickness, so its efficiency is predictably less than that of the other detectors. However, its excellent resolution (compared with other portable detectors) offsets lower efficiency in some cases.

Using the four detectors, we also measured plutonium sources. Figure 3 shows the spectra of low-burnup plutonium taken with the four detectors.

Although the resolution measured with the CdTe detector is worse than that of the Ge, it is much better than that of the NaI and CdZnTe detectors. Many closely spaced peaks (such as the 203-keV and 208-keV lines) not resolved with NaI or CdZnTe are completely or partially resolved in the CdTe spectra. This resolution extends the "reach" of higher resolution spectroscopy to *in-situ* field applications of NDA for safeguards and nonproliferation. Wide-range plutonium isotopic analysis is now possible using gamma-ray spectra measured with CdTe.

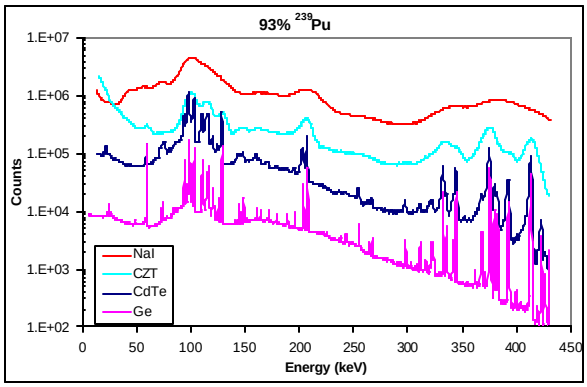


**Figure 3. Gamma-ray spectra of low-burnup plutonium measured with the four detectors. The spectra are shifted vertically for clarity.**

**B. CdTe Detector Stability and Linearity**

Knowledge of the stability and linearity of the CdTe detector is required to develop it as a useful tool. Detailed measurements of these performance characteristics are described elsewhere.[3] The large integral nonlinearity will complicate the plutonium isotopics analysis somewhat.

**C. Plutonium Isotopic Results with First CdTe**

Plutonium and uranium isotopic analysis of gamma-ray spectra taken by Ge was the original purpose of FRAM. It is unique because of a sophisticated external parameter database structure. Applications to all high-resolution gamma-ray detector types, element/isotope combinations, gamma-ray-energy ranges, and interference peaks are accessible from the parameter file of FRAM, unlike other isotopic tools for which new versions must be generated to address many of these differences. This somewhat mechanistic distinction between FRAM and other isotopic analysis programs is responsible in large part for its success in analyzing the CdTe plutonium spectra.

There are several differences between CdTe and Ge spectra. These differences must be addressed to successfully do isotopic measurements with CdTe. Even though the resolution of the CdTe detector is excellent compared to that of other portable detector systems, it is still about three times worse than that of Ge. Some peaks that are clearly separated in a Ge spectrum may not be completely resolved in a CdTe spectrum. This is a challenge for extracting information from such peaks. Also, broader peaks greatly reduce the regions available for determining the continuum background for subtraction from the peaks.

Another challenge is the peak shape. The peak shapes for CdTe exhibit three different tailing components: one low-energy short-term, one low-energy long-term, and one high-energy short-term. Note that for Ge, the low-energy long-term and the high-energy short-term tails are nonexistent or very small and can be ignored. These tails are large for CdTe, different for different energies or shaping times, and dependent on operation time.

FRAM fits a background-subtracted peak with a Gaussian function plus a low-energy exponential tailing function. This is not sufficient for the CdTe peaks, but the results are still quite good for a hand-held, noncryogenic system.

Using the CdTe detector, we measured 13 small reference samples of plutonium oxide of well known isotopic composition covering a wide range of burnup, from a low $^{240}$Pu fraction of 3.6% to a high of 26% with masses ranging from 0.4 to 20 g.

We used three different MCA systems (DSPEC Plus, Inspector 2000, and MCA-166) to collect data from these 13 samples. The acquisition time was one hour for each spectrum. Twenty-one spectra were obtained.

We modified FRAM v4 to account for broad peaks with large tails and also introduced a generic CdTe detector efficiency curve into the FRAM code. The CdTe spectra were then analyzed. The biases for all the samples appear to be randomly distributed with no apparent trend. The average results of all 21 spectra with the 13 samples are shown in Table 3.

**Table 3. Bias in FRAM analysis of reference gamma-ray spectra obtained with the first CdTe detector. % bias = 100·(Meas. – Ref.)/Ref.**

| | $^{238}$Pu | $^{239}$Pu | $^{240}$Pu | $^{241}$Pu | $^{241}$Am |
|---|---|---|---|---|---|
| **Ave bias** | -7.10 | -0.54 | 2.99 | 0.12 | -2.23 |
| **%STDEV** | 21.78 | 1.56 | 8.12 | 4.84 | 8.11 |

**III. PERFORMANCE: SECOND CdTe DETECTOR**

We learned from the performance of the first detector that the plutonium isotopic analysis with CdTe can be improved by

- better detector (efficiency, resolution, and peak shape).
- better MCA (or the MCA can be better optimized for the CdTe detector).

- better peak-fitting software that can address the three tailing components of a peak.
- extending the analysis energy regions to exploit larger peak intensities than those between 125 keV and 414 keV.

We improved the CdTe plutonium isotopic analysis with the second detector by applying the improvements shown in the first three bulleted items above.

This second detector is slightly larger than the first one with dimensions of 11 mm x 9 mm x 2.1 mm. Its efficiency in the energy range from 125 to 414 keV is about 25% better than that of the first detector. Its nominal resolution of 1.6 keV at 122 keV is also better than that of the first detector.

Given more time to optimize the second detector and the DigiDART MCA, we were able to improve both the resolution and peak shape. The result is that the code FRAM, even with only one tailing component, gives an improved fit to the peaks of this detector. We also slightly modified the way the code determines the tails of the CdTe peaks, which improves the final results.

We obtained data for the four 6-g plutonium reference sources with $^{240}$Pu fractions of 6, 14, 19, and 26%, respectively. We acquired multiple 1-h spectra, ranging from 23 to 27 spectra, with each source. One hundred spectra were obtained. The data were analyzed using the improved code. The results are shown in Table 4.

**Table 4. Bias in FRAM analysis of reference gamma-ray spectra obtained with the second CdTe detector. % bias = 100·(Meas. – Ref.)/Ref.**

| | $^{238}$Pu | $^{239}$Pu | $^{240}$Pu | $^{241}$Pu | $^{241}$Am |
|---|---|---|---|---|---|
| **Ave bias** | 0.45 | 0.37 | -0.67 | -2.31 | 0.88 |
| **%STDEV** | 6.99 | 0.81 | 4.24 | 2.93 | 6.85 |

## IV. DISCUSSION

The reported errors from FRAM arise mainly from the statistics. For the first detector, the standard deviation of the bias divided by reported error values of $^{240}$Pu is 2.2. (For the Ge detector, this value is typically about 1.) The larger bias for CdTe is mainly a result of the imperfect fit of Ge peak shapes to CdTe peaks. Better peak-fitting routines or better peak shapes will improve the results with the first detector by as much as a factor of 2.2. This is demonstrated with the second detector whose results are about a factor of 2 better than those of the first. Some of this factor of 2 is caused by the higher efficiency of the second detector and better resolution, but most of the improvement comes from better fits of the peaks.

The new Peltier-cooled CdTe detectors are commercially available at a cost that is between that of NaI and Ge detectors (and comparable to the cost of CdZnTe detectors with similar crystal dimensions). Larger and thicker CdTe detectors may eventually replace CdZnTe and NaI detectors in many applications that require portable systems. Because the performance of the CdTe detector is much improved over that of CdZnTe, the CdTe crystals required to replace CdZnTe can be smaller than those of CdZnTe. The CdTe detectors should replace Ge detectors in some applications that require good resolution. An example is the wide-range gamma-ray isotopics analysis of plutonium described in this paper. Portable gamma-ray isotopics measurements of *in-situ* plutonium can be addressed by using the present CdTe detectors, whose crystals are sufficiently large to achieve the required isotopic results.

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